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Stochastic Analysis and Control in Kinetics of Multistable Chemical Reactor

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Abstract: We consider a model of thermochemical reactor proposed by Nowakowski. Stochastic effects in the bistability zone are studied. A parametric analysis of noise-induced transitions between coexisting equilibria is carried out on the basis of the stochastic sensitivity technique and confidence ellipses method. We solve the problem of stabilization of the equilibrium regime under incomplete information. The feedback regulator which reduces the stochastic sensitivity and stabilizes the randomly forced equilibrium is constructed.

Keywords: Thermochemical model, noise-induced transitions, feedback regulator, stochastic sensitivity, stabilization.

1. INTRODUCTION

In real processes, an interplay of the nonlinearity and stochasticity can essentially complicate the dynamic behavior. Even a weak noise in strongly nonlinear systems generates the wide variety of stochastic phenomena (see, e.g. Anishchenko et al. (2007); Horsthemke (1984); Fedotov et al. (2002, 2006)). Complex dynamic regimes observed in the mathematical models of chemical kinetics (Field et al. (1985); Jakobsen (2014)) are extremely sensitive to noise.

In the present paper, we study the nonlinear model of thermochemical reactor proposed in (Nowakowski et al. (2005); Kawczyński et al. (2008)). This multistable model demonstrates a diversity of dynamic regimes. We show that the deterministic stability can be insufficient for the proper operation of the flow reactor in the presence of inevitable noise. For the study of the probabilistic mechanisms of the noise-induced transitions, we apply the stochastic sensitivity analysis (Bashkirtseva et al. (2010); Ryashko (2018)).

To provide a proper operation of stochastically forced systems, we develop an appropriate control approach. Now, control theory of nonlinear stochastic systems is actively developed (see, for instance, Kushner (1967); Astrom (1970); Sun (2006); Panteleev et al. (2018); Azanov et al. (2018); Pakshin et al. (2016) and bibliography therein). A control method based on the stochastic sensitivity synthesis was developed in (Ryashko et al. (2008); Bashkirtseva et al. (2012, 2017)).

In the present paper, on the example of the Nowakovski model, we show how this mathematical technique can be used in the solution of the important engineering problem of the stabilization of the flow chemical reactor under incomplete information.

2. DETERMINISTIC MODEL

Consider a model (Nowakowski et al. (2005)) of the thermochemical reaction in the flow reactor with well mixing:

$$
\begin{aligned}
\dot{x} &= \sqrt{y}\left(-x\exp\left(-\frac{\delta}{y}\right)+p\,(1-x)\right)\\
\dot{y} &= \frac{2}{3}q\sqrt{y}\left(x\exp\left(-\frac{\delta}{y}\right)+r\,(1-y)\right)
\end{aligned}. \qquad (1)
$$

Here, the variable x is the concentration of the reagent, and y is a temperature inside the reactor. The parameters p, q, r and δ are positive.

This model exhibits a variety of complex dynamic regimes, both mono- and bistable. In the present paper, we focus on the bistable regime with two stable equilibria. Following Nowakowski et al. (2005), we fix $\delta = 5$, $r = 0.07$, $q = 40$, $p = 0.5$.

For this set of parameters, the phase portrait and corresponding time series are plotted in Fig. 1. As one can see, system (1) possesses two stable equilibria $M_1(0.7544, 2.7544)$ and $M_2(0.9634, 1.2612)$, separated by the saddle equilibrium $M_3(0.8719, 1.9153)$. The basins of attraction of equilibria M_1 and M_2 are separated by the stable manifold of the saddle M_3.

As can be seen, a kinetics of the chemical reaction essentially depends on the initial values of the concentration and temperature. Depending on the initial data, the reaction is stabilized either to the equilibrium M_1 or to the equilibrium M_2.

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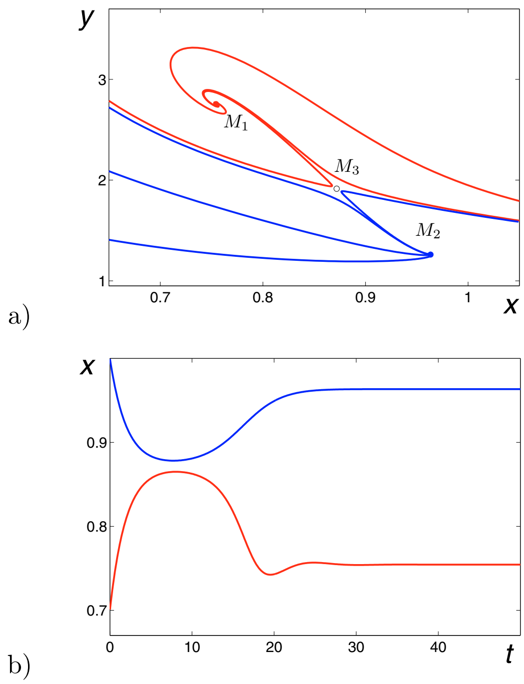


Fig. 1. Deterministic system with $\delta = 5,\ r = 0.07,\ q = 40,\ p = 0.5$: a) phase portrait, b) time series.

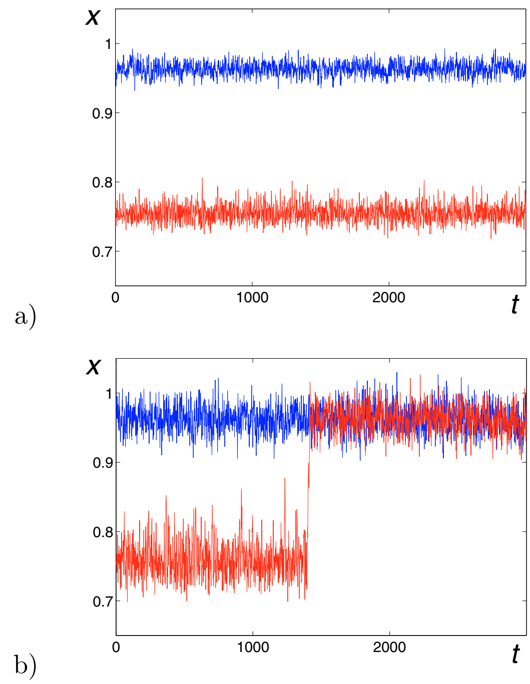


Fig. 2. Stochastic system with $\delta = 5,\ r = 0.07,\ q = 40,\ p = 0.5$: a) for $\varepsilon = 0.01$, b) for $\varepsilon = 0.02$.

3. STOCHASTIC MODEL

To study an influence of the inevitable random forcing on the kinetics of the thermochemical processes, we will use the following stochastic model:

$$\begin{aligned} \dot{x} &= \sqrt{y}\left(-x\exp\left(-\frac{\delta}{y}\right)+p\,(1-x)\right)+\varepsilon_1\xi_1(t),\\ \dot{y} &= \frac{2}{3}q\sqrt{y}\left(x\exp\left(-\frac{\delta}{y}\right)+r\,(1-y)\right)+\varepsilon_2\xi_2(t). \end{aligned} \tag{2}$$

Here, $\xi_{1,2}(t)$ are standard Gaussian uncorrelated noises, and $\varepsilon_{1,2}$ are noise intensities. It is supposed that $\varepsilon_1 = \varepsilon_2 = \varepsilon$.

In Figure (2), time series of x-coordinates of solutions of system (2) are plotted for two values of the noise intensity. By red color, we show trajectories starting from M_1, and by blue color, we show trajectories starting from M_2.

For weak noise $\varepsilon = 0.01$, stochastic trajectories starting from the equilibria M_1 and M_2 oscillate with small amplitude near the initial deterministic attractors (see Fig. 1a). When the noise intensity increases, a transition from the basin of attraction of M_1 to M_2 occurs (see Fig. 2b for $\varepsilon = 0.02$). So, the noise transits the considered thermochemical system to another alternative noisy equilibrium regime. From the Fig. (2), one can see that the equilibrium M_1 is more sensitive to noise than M_2. This can be confirmed quantitatively by the stochastic sensitivity analysis (see Section5).

For equilibria M_1 and M_2, one can find corresponding stochastic sensitivity matrices W_1 and W_2. Let us compare their eigenvalues. For W_1, we have $\lambda_1 = 100.302,\ \lambda_2 = 0.818$. For W_2, we have $\lambda_1 = 1.987,\ \lambda_2 = 0.708$. So, the equilibrium M_1 is much more sensitive to noise.

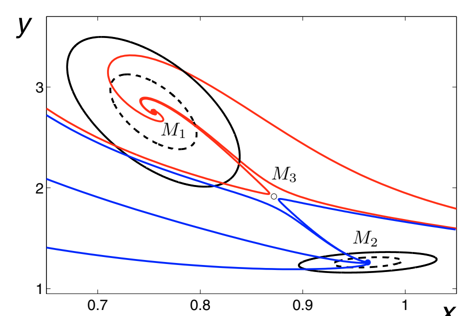


Fig. 3. Confidence ellipses for $\delta = 5,\ r = 0.07,\ q = 40,\ p = 0.5$ and $\varepsilon = 0.01$ (dashed), $\varepsilon = 0.02$ (solid). Here, the fiducial probability $\mathcal{P} = 0.999$.

The scenario of the transition from M_1 to M_2 can be analysed with the help of confidence ellipses (see (9)). In Figure (8), we plot confidence ellipses around M_1 and M_2 for $\varepsilon = 0.01$ by dashed lines, and for $\varepsilon = 0.02$ by solid lines.

Consider ellipses around M_1. For weak noise ($\varepsilon = 0.01$), the confidence ellipse totally belongs to the basin of attraction of M_1. For larger noise ($\varepsilon = 0.02$), the confidence ellipse expands, and begins to occupy the basin of attraction of M_2. This signals about the possible noise-induced transition of random trajectories from M_1 to M_2. Note that both ellipses around M_2 lie inside the basin of attraction of M_2, so, random trajectories starting from M_2 are still near M_2.

As one can see, results of the stochastic sensitivity analysis well agree with direct numerical simulations.

Note that such noise-induced transitions may be unacceptable from the engineering point of view. Often, it is

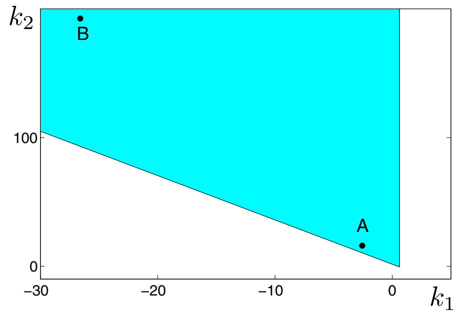


Fig. 4. The stability domain $\mathbf{K}$ for $\delta = 5, r = 0.07, q = 40, p = 0.5$.

required to ensure the functioning of the system near a given equilibrium regime. In the next Section, we consider how with the help og the feedback regulator one can reduce a stochastic sensitivity of the equilibrium and stabilize the normal operating mode of the thermochemical system.

4. CONTROLLING STOCHASTIC SYSTEM

Consider the thermochemical stochastic model with control inputs u_1 and u_2:

$$\begin{aligned}\dot{x} &= \sqrt{y}\left(-x\exp\left(-\frac{\delta}{y}\right)+p\,(1-x)\right)+u_1+\varepsilon_1\xi_1(t),\\ \dot{y} &= \frac{2}{3}q\sqrt{y}\left(x\exp\left(-\frac{\delta}{y}\right)+r\,(1-y)\right)+u_2+\varepsilon_2\xi_2(t).\end{aligned} \tag{3}$$

To demonstrate abilities of our control technique, we will consider a case of the incomplete information, when only the coordinate x can be measured. In these circumstances, to stabilize the system (3) near the equilibrium $M_1(\bar{x},\bar{y})$, we will use the feedback regulator

$$u_1 = k_1(x-\bar{x}), \qquad u_2 = k_2(x-\bar{x}). \tag{4}$$

In this case (see general formulas in Section 5),

$$K = \begin{bmatrix} k_1 \\ k_2 \end{bmatrix}, \quad C = [\,1\;\;0\,], \quad F+BKC = \begin{bmatrix} f_{11}+k_1 & f_{12} \\ f_{21}+k_2 & f_{22} \end{bmatrix}.$$

Here,

$$F = \begin{bmatrix} f_{11} & f_{12} \\ f_{21} & f_{22} \end{bmatrix}$$

is the Jacobi matrix for the equilibrium M_1 in the deterministic system (1). The set $\mathbf{K}$ of the regulator (4) parameters guaranteeing the exponential stability of the equilibrium M_1 is defined by the system of linear inequalities:

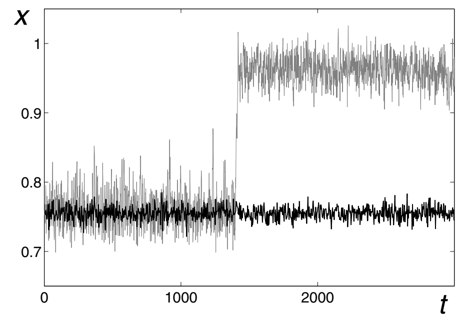
$$\begin{aligned}&\mathrm{tr}(F+BKC) = f_{11}+f_{22}+k_1 < 0,\\ &\det(F+BKC) = f_{11}f_{22}-f_{12}f_{21}+f_{22}k_1-f_{12}k_2 > 0.\end{aligned} \tag{5}$$

This stability domain $\mathbf{K}$ is shown in Fig. 4.

For any $(k_1,k_2)\in\mathbf{K}$, elements $w_{ij}(k_1,k_2)$ of the stochastic sensitivity matrix $W(k_1,k_2)$ can be found from the system (8). The problem of the reducing the sensitivity can be solved by the standard descent procedures.

For example, for $k_1 = -2.56,\ k_2 = 15.97$ (see the point A in Fig. 4), eigenvalues of the stochastic sensitivity matrix are

$$\lambda_1 = 64.78,\ \lambda_2 = 0.158.$$



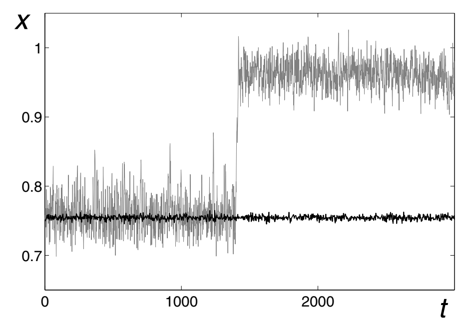


Fig. 5. Time series of stochastic system (3) with $\delta = 5,\ r = 0.07,\ q = 40,\ p = 0.5,\ \varepsilon = 0.2$ without control (grey), and with regulator with parameters k_1 and k_2 corresponding to points a) A, b) B from the Fig. 4.

For $k_1 = -26.6,\ k_2 = 192.5$ (see the point B in Fig. 4) we have

$$\lambda_1 = 55.7,\ \lambda_2 = 0.018.$$

Remember, that for this system without control, for M_1 we have $\lambda_1 = 100.302,\ \lambda_2 = 0.818$.

As can be seen, the regulator (4) with presented coefficients k_1, k_2 can decrease the stochastic sensitivity of M_1. Results of the numerical simulation of the system (3) with regulator (4) are shown in Fig. 5.

It is worth noting that the suggested control approach allows us to stabilize the required equilibrium regime of the stochastic thermochemical reactor and suppress unwanted noise-induced transitions.

5. CONTROL OF STOCHASTIC EQUILIBRIUM UNDER INCOMPLETE INFORMATION

Consider a stochastic system

$$\dot{x} = f(x,u)+\varepsilon\sigma(x)\xi(t) \tag{6}$$

with the regulator

$$u = K(y-\bar{y}), \quad y = g(x), \quad \bar{y} = g(\bar{x}). \tag{7}$$

Here, x is an n-dimensional state, u is an l-dimensional control, $f(x,u)$ is an n-vector-function, $\sigma(x)$ is and $n\times q$-matrix-function, $\xi(t)$ is an q-dimensional standard Gaussian process with parameters $\mathrm{E}\xi(t) = 0,\ \ \mathrm{E}\xi(t)\xi^\top(\tau) = \delta(t-\tau)I$ (I is the identity matrix), and ε is the noise intensity.

In the regulator (7), we use the measurement m-vector $y(t)$ and the equilibrium $\bar{x}$ of the system (6) with $\varepsilon = 0$, $u = 0$. The stability of $\bar{x}$ is not assumed. The regulator (7) is used when the available information on the current state $x(t)$ is incomplete. In (7), the $l \times m$-matrix K is constant.

Denote by $\mathbf{K}$ a set of matrices K that ensure the exponential stability of the equilibrium $\bar{x}$ for the system (6),(7) for $\varepsilon = 0$. This set $\mathbf{K}$ can be described as

$$\mathbf{K} = \{K |\ Re\lambda_i(F + BKC) < 0\},$$

where

$$F = \frac{\partial f}{\partial x}(\bar{x}, 0), \quad B = \frac{\partial f}{\partial u}(\bar{x}, 0), \quad C = \frac{\partial g}{\partial x}(\bar{x}),$$

and $\lambda_i(F+BKC)$ are eigenvalues of the matrix $F+BKC$.

For any $K \in \mathbf{K}$, the sensitivity of the equilibrium $\bar{x}$ to noise in the system (6),(7) is defined by the stochastic sensitivity matrix W Ryashko et al. (2008). This matrix satisfies the following matrix equation

$$\begin{aligned} &(F + BKC)W + W(F + BKC)^\top + S = 0, \\ &S = \sigma(\bar{x})\sigma^\top(\bar{x}). \end{aligned} \tag{8}$$

Using the stochastic sensitivity matrix W one can find the first approximation of the covariance matrix of the stationary distributed solutions $\bar{x}^\varepsilon(t)$ of system (6),(7):

$$\mathrm{cov}(\bar{x}^\varepsilon(t), \bar{x}^\varepsilon(t)) \approx \varepsilon^2 W.$$

For 2D-system, eigenvalues $\lambda_1,\ \lambda_2$ and eigenvectors $v_1,\ v_2$ of the matrix W define the confidence ellipse

$$\frac{z_1^2}{\lambda_1} + \frac{z_2^2}{\lambda_2} \ = \ 2\varepsilon^2 q, \quad q = -\ln(1 - \mathcal{P}). \tag{9}$$

Here, $\mathcal{P}$ is a fiducial probability, $z_i = (x - \bar{x}, v_i),\ i = 1, 2$.

So, to control the dispersion of random states around the equilibrium $\bar{x}$, one has to synthesize the required stochastic sensitivity matrix W by the appropriate regulator (7). For any $K \in \mathbf{K}$, the regulator (7) forms the corresponding stochastic sensitivity matrix W_K of the equilibrium $\bar{x}$. Let $\mathbf{M}$ be a set of symmetric and positive definite $n \times n$-matrices. For the assigned matrix $W \in \mathbf{M}$, one has to find a matrix $K \in \mathbf{K}$ guaranteeing the equality $W_K = W$, where W_K is a solution of equation (8).

As a result, the problem of the synthesis of the assigned stochastic sensitivity matrix W is reduced to the search of the matrix K satisfying the matrix equation

$$\begin{aligned} &BKCW + WC^\top K^\top B^\top + H(W) = 0, \\ &H(W) = FW + WF^\top + S. \end{aligned} \tag{10}$$

The equation (10) is equivalent to the equation

$$BKC = (Q - FW - 0.5S)W^{-1}, \tag{11}$$

where Q is an arbitrary skew-symmetric $n \times n$-matrix.

In non-singular case, the matrix K of the regulator (7) which synthesizes the required matrix $W \in \mathbf{M}$, the solution of the equation (11) can be found as

$$K = B^{-1} [Q - FW - 0.5S] W^{-1} C^{-1}. \tag{12}$$

So, the regulator (7) with the feedback matrix (12) can synthesize any required stochastic sensitivity matrix.

In the general case, the solution of equation (11) requires the pseudo-inversion theory Albert (1972). Here, additional conditions of the solvability of (11) are as follows:

$$\begin{aligned} &(I - BB^+) [Q - FW - 0.5S] = 0, \\ &[Q - FW - 0.5S] W^{-1}(I - C^+C) = 0. \end{aligned} \tag{13}$$

Under these conditions, the equation (11) has a solution

$$K = B^+ [Q - FW - 0.5S] W^{-1} C^+. \tag{14}$$

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